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# RADON PROGENY AND THORON PROGENY PLATE-OUT ON A VARIETY OF MATERIALS

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# RADON PROGENY AND THORON PROGENY PLATE-OUT ON A VARIETY OF MATERIALS

by

J. Bigu* and A. Frattini**

## ABSTRACT

The radon and thoron progeny plate-out characteristics of a variety of materials have been investigated in a large (26 $m^3$) radon/thoron test facility (RTTF). Discs (about 25 mm diameter) were made of different materials including metals, fabrics, plastics and filter materials. The discs were placed on the walls of the RTTF for at least 24 hours. After exposure, the materials were removed from the RTTF and their surface α-activity was measured. Significant differences in the surface α-activity were found between different materials. Experiments were conducted preferentially at low relative humidity and low aerosol concentrations. The plate-out differences between the various materials ranged from less than 10% to over 60%. This work is relevant to the determination of deposition velocities on, and attachment rates to, large surfaces.

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# L'hadérence des produits de filiation du radon et du thoron à la surface de divers matériaux

par

J. Bigu* et A. Frattini**

## RÉSUMÉ

Les caractéristiques d'adhérence en surface des produits de filiation du radon et du thoron sur divers matérieaux ont été analysées dans une grande installation d'essai du radon et du thoron (26 $m^3$). Les disques utilisés (environ 25 mm de diamètre) étaient composés de différents matériaux dont le métal, le tissu, le plastique et les matériaux pour filtres. Ces disques ont été placés sur les parois de l'installation mentionnée pendant au moins 24 heures. Les matériaux ont ensuite été retirés de l'installation et l'activité alpha de leur surface a été analysée. Cette activité a accusé des variations significatives entre les différents matériaux. Les expériences ont été réalisées de préférence dans des conditions de faible humidité relative et de faible concentration d'aérosols. Les différences d'adhérence entre les différents matériaux ont varié de moins de 10 % à plus de 60 %. Ce projet s'insère dans la détermination des vitesses de dépôt et de fixation sur les grandes surfaces.

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# INTRODUCTION

The short-lived decay products of radon and thoron are initially formed in an atomic, positively charged, state of great diffusivity. These decay products readily attach themselves to small surfaces such as aerosols, and to large surfaces such as walls. Attachment of particles to large surfaces is commonly referred to as plate-out. The degree of plate-out depends on environmental conditions including aerosol concentration, air moisture content, the presence of trace gases, and most probably, on the properties of the surface. Although theoretical and experimental data are available on the attachment of the radon and thoron progeny to aerosols, less is known on the attachment to walls, with regards to the chemical nature and physical characteristics of the wall surface.

Plate-out of the radon and thoron progeny on walls and other objects is a problem of great practical interest and concern when evaluating radiation levels by environmental monitors and personal dosimeters. This is so because plate-out affects the accuracy and reliability of the results obtained by some radiation instrumentation as decay products of radon and thoron may be removed by the 'sampling heads' and other components of the instrument. Plate-out of these decay products is also relevant in connection with air quality control in underground uranium mines, mills, and other uranium-related industries, and in the operation of radon (thoron) test facilities for simulation studies, calibration of instrumentation and for other practical purposes.

Data on the rate of attachment (plate-out) of radioactive particles on walls are used to calculate the particle deposition velocity. Both variables i.e., attachment rate and deposition velocity are used in turn to estimate the extent of plate-out on sampling heads, and to evaluate the effect of large surfaces on radiation levels in specific working areas and test facilities, as

indicated above.

Deposition rates are determined experimentally by measuring the surface activity on some samples designed for the purpose. These samples usually assume the form of small 'discs' which are attached to the surface where plate-out is to be determined. The samples are used because plate-out measurements carried out directly on the surface of interest may not be practical, and in most cases quite difficult.

Furthermore, because of practical considerations, a limited number of materials are used for this purpose under the assumption that the mechanism, or extent, of deposition is independent of the type of material used. As there is little evidence to support or deny this view, work in this area is relevant. It should be noted that materials commonly used in plate-out experiments to determine attachment rates and deposition velocities such as metals and filter paper, may not necessarily be the most representative ones for the purpose, as the characteristics of these materials may differ significantly from the properties of the surfaces from where data are required. If the mechanism, or extent, of particle deposition is somewhat dependent on the material used and/or the properties of its surface, data obtained by employing certain materials may not reflect true experimental conditions, and therefore, lead to significant errors in the estimation of plate-out effects.

This paper deals with the plate-out characteristics of a variety of materials, including metals, plastics, some fabrics and powders to the decay products of radon and thoron under laboratory-controlled conditions. In a previous paper, the author reported on measurements on the attachment rate and deposition velocity of radon and thoron decay products (1). In these experiments, stainless steel discs and filter paper were used. At the time, the assumption was made that the attachment of the decay products was

independent of the chemical and physical nature of the surface on which the products were deposited. The present work sets out to verify this contention.

## EXPERIMENTAL PROCEDURE

Experiments were conducted in a large radon/thoron test facility (RTTF) designed for calibration purposes and simulation studies (2). A number of different materials were exposed in the RTTF to a radon/radon daughter or thoron/thoron daughter atmosphere. Exposure of the materials was carried out under laboratory-controlled conditions of radiation level, aerosol concentration, air moisture content and temperature.

The materials used were in the form of circular 'discs' of the same thickness ($\sim$0.5 mm) and diameter ($\sim$25 mm). These were placed at different locations on the walls of the RTTF at about 1.6 m above the floor. Samples (discs) of different materials were arranged in sets of 3 to 4; they were placed close to one another to ensure exposure under identical conditions. Exposure time was at least 24 hours to ensure surface activity equilibrium conditions. After a given exposure time, the discs were removed from the RTTF and their surface $\alpha$-activity was measured as a function of time, up to 2 hours, using standard radiation instrumentation and methods. Counting of samples began 2 min after their removal from the RTTF.

For the thoron progeny, samples were counted for 15 min followed by 5 min counts with 1 min interval between counts. For the radon progeny two different counting routines were used, namely: 2-min counts with 1-min interval, and 5-min counts with 1-min interval. The latter counting routine was intended for more precise $^{218}Po$ measurements.

The surface activity on the samples for each exposed set of samples was compared. Each set of samples was exposed, and measured, a minimum of about five times to improve overall statistics. Experiments were conducted in the

temperature range 19-22°C and at low relative humidity (10 to <40%). The aerosol concentration, mainly natural atmospheric aerosols, was kept low, i.e., $5 \times 10^2$ to $9 \times 10^3$ $cm^{-3}$. No attempt was made to maintain the aerosol concentration at any particular level at these low aerosol concentrations. The values attained were those due to controlled unfiltered air, purposely leaking into the test facility used for the experiments. The aerosol concentration varied with barometric pressure. The conditions of low relative humidity and low aerosol concentration is of great practical interest because plate-out phenomena are substantially increased under these conditions.

The materials investigated included several metals (copper, Cu; aluminum, Al; stainless steel, SS; and galvanized steel, GS); filter materials (Glass Fiber, Millipore and Varsopor); plastic materials (Plexiglas); 'powders' (activated carbon, talcum); and other materials such as sandpaper, emery cloth and several fabrics. Activated carbon and talcum powder samples were prepared by depositing a thin layer (~0.5 mm) of the powder on filter paper on which some binding material (glue) had been added to its surface to retain the powder.

## RESULTS AND DISCUSSION

The results obtained in these series of measurements are shown in Figures 1 to 11. Figure 12 shows α-spectra taken from a copper disc exposed in the RTTF to a radon progeny atmosphere (α-spectrum on the right), and a thoron progeny atmosphere (α-spectrum on the left) for about 24 hours.

Figures 1 to 4 show the surface α-activity measured on several materials from exposure to a radon progeny atmosphere. Significant differences on the surface α-activity were found between some of the materials investigated. Under our experimental conditions, the following was observed; for radon progeny atmospheres, plate-out on Al was greater than for Cu, which

in turn was greater than for GS. Plate-out on Plexiglas was greater than for Cu and Al. Plate-out on Glass Fiber filters was greater than for some fabrics, sandpaper and emery cloth.

As expected, Figures 1 to 4 show that the surface α-activity on the materials examined decreased with increasing time after exposure. The approximate half-life corresponding to the surface α-activity was in the range of 35 to 50-min. These values roughly agree with the 'combined' half-life of the short-lived decay products of radon.

The differences in the decay curves for different materials and for a given material under different environmental conditions, i.e., radiation level, temperature, relative humidity and aerosol concentration are mainly due to the different radioisotope make-up about the surface of the materials investigated. This radioisotope make-up very much depends on the radon progeny disequilibrium ratios, i.e., $[^{214}Pb]/[^{218}Po]$ and $[^{214}Bi]/[^{218}Po]$, where the square brackets are used to denote activity concentration. The radon progeny disequilibrium ratios are highly dependent on the air flow characteristics, relative humidity and aerosol concentration in the RTTF.

Figures 1 to 4 also show a rapid decrease in α-activity during the first few minutes after removal of the samples from the RTTF followed by a less pronounced decrease in the activity. This behaviour is due to the rapid decay of $^{218}Po$ with a half-life of 3.05 min. The lack of smoothness in some of the graphs presented here is attributed to statistics of counting. A better understanding of plate-out phenomena can be gained by a theoretical study on the growth and/or decay of activity plated-out on the samples during and after the exposure period. This problem has been dealt with by Bigu elsewhere (3).

Figures 5 to 11 show the surface α-activity measured on several materials from exposure to a thoron progeny atmosphere. As for the case of the radon

progeny, quite substantial differences in the surface α-activity were found between several of the materials studied. Differences for the thoron progeny appear to be more accentuated, under our experimental conditions, than for the radon progeny. In general terms, substantial plate-out differences were found between Plexiglas and Cu; between GS and Cu; and between Al and Cu (see Figures 5 and 6). No significant difference was found between GS, Plexiglas and Al (see Figure 7). Quite significant differences were found between some fabrics, fiberglass material (filters), emery cloth, membrane materials (e.g., Millipore filters) and medium steel sandpaper (MSS), (see Figures 8, 9 and 11), and between some powders (e.g., talcum and activated carbon) thinly spread over a paper substrate and a 'plain' paper sample (see Figure 10).

Because of practical limitations in the number of α-scalers (i.e., 3) available at the time of experimentation and the difficulty in maintaining the radon (thoron) progeny level for extended periods of time (i.e., 3 months) within less than $\pm$8% of a given level, no direct comparison between all the materials investigated, except for three materials at a time, can be made in order to rank their plate-out characteristics. Experiments were intentionally conducted at different radiation levels suitable for other specific purposes, including dosimetric studies.

The plate-out differences between the materials investigated here ranged from less than 10% to over 60% (see Figures 1 to 11).

Figures 5 to 11 show that the surface thoron progeny α-activity remained fairly constant during the counting period, i.e., about 100-min. This result indicates that the α-activity measured on the surface of the materials was mainly due to Bi-212, and Po-212 in equilibrium with Bi-212, which were in turn in equilibrium with the relatively long-lived, 10.6 hour half-life, Pb-212. Hence, Bi-212 and Po-212 decayed with the half-life of Pb-212.

Figure 12 shows sharp α-spectra for the radon and thoron progeny deposited on samples. These sharp spectra are to be expected from non-dusty atmospheres and passive sampling.

The data presented here suggest some qualitative and/or quantitative differences in behaviour between the radon progeny and thoron progeny relative to some materials, and for some materials relative to either the radon progeny or the thoron progeny.

Other data, not presented here, also suggest that the plate-out characteristics of some materials depend on air moisture content, aerosol and dust concentration and size distribution and other variables in a rather complex, not clearly understood, fashion. Hence, substantial differences may arise when the same set of materials are investigated under different environmental conditions.

At present, only some speculation can be offered regarding the cause for the observed plate-out characteristics of different materials. The following factors, or a combination of factors, may be relevant to the present discussion:

1. Electrostatic charge on the surface of the material. Air flow characteristics may affect deposition of charge.
2. Short-range and long-range surface forces.
3. Chemical composition and physical state and structure of the material surface.
4. Radon progeny and thoron progeny chemical affinity, i.e., of Po, Bi and Pb radioisotopes, with the material.
5. Effect of air moisture, trace gases and aerosol composition, which may lead to chemical reactions with the radon and thoron progeny and/or the material. These chemical reactions could affect the plate-out characteristics of the material (see item 4).

6. Radon and thoron progeny desorption by recoil processes.
7. 'Roughness' of the surface of the material which can affect the mass transfer characteristics between the radon and thoron progeny and the surface of the material. Mass transfer processes depend on a number of factors, including air flow characteristics and the geometry, i.e., shape, dimensions, and surface characteristics, of the material.
8. Effective surface area of the material surface, as opposed to 'macroscopic' geometrical surface area calculated according to, say, the diameter of the disc. The effective surface area of the material is partly determined by 'irregularities' in the distribution of the material, at the atomic and molecular levels, on its surface. The effective surface area could conceivably be much larger than the cross-sectional area based on purely geometrical considerations.

There is very little information available in the literature on the effect of items 2) to 6) on the radon and thoron progeny plate-out characteristics of different materials. The effect of trace gases on the diffusion coefficient of the unattached radon progeny has been reviewed recently by Busigin et al. (4). A review of the literature covering some aspects of item 7) and other considerations relevant in the context of this paper has been published recently by Porstendorfer (5). See also references 6 and 7.

The effect of surface electrostatic charge on a material on the attachment of the decay products of radon has been known since pioneering work on atomic structure by Rutherford. Extensive research into this area for the radon and thoron progeny has been conducted in this laboratory for environmental monitoring purposes. Card and Bell reported on the effect of electrostatic charge on the collecting characteristics of copper for the radon progeny for exploration purposes (8).

In a series of recent thoron progeny experiments, dramatic differences in the α-activity collected on copper discs were observed by the authors when a positive charge, a negative charge, or a zero charge, i.e., discs at ground potential, was applied. Discs positively charged (∿+270 V) collected 1.4 to 2.8 (ave. : 2.0) times more α-activity than discs at zero potential. Discs negatively charged (∿-270 V) collected 11 to 21 (ave. : 17) times more α-activity than discs at zero potential. The α-activity ratios between discs charged at -270 V and +270 V were in the range 4.5 to 15.5 (ave. 9.5). The above figures, and figures for the radon progeny, obtained in the past, varied quite substantially with environmental conditions in a rather complex and little understood fashion.

The effect of item 8) is at present under investigation using electron-microscopy techniques.

## CONCLUSIONS

Significant differences in the radon and thoron progeny plate-out characteristics of different materials have been found. As radon and thoron progeny deposition velocities are normally determined by using some material as a collector of activity, the choice of collecting surface is important in view of our results. The arbitrary choice of a material as an 'activity' collector may lead to substantial errors in the determination of deposition velocities, and other variables, if the data obtained for this particular material are to be used as representative of the deposition velocity on a large surface made of a different material. It should be noted that there are some practical difficulties in determining radon and thoron progeny activity directly on the surface of interest.

Some of the data reported here, and unpublished data, seem to indicate that there might be some qualitative difference between the behaviour of the

radon progeny and thoron progeny towards a given material.

The underlying physical and/or chemical mechanisms responsible for the differences observed between the radon progeny and the thoron progeny as related to different materials are not clearly understood, but it is presumed to involve one or more of items 1) to 8) indicated above. Much more research is needed on plate-out phenomena.

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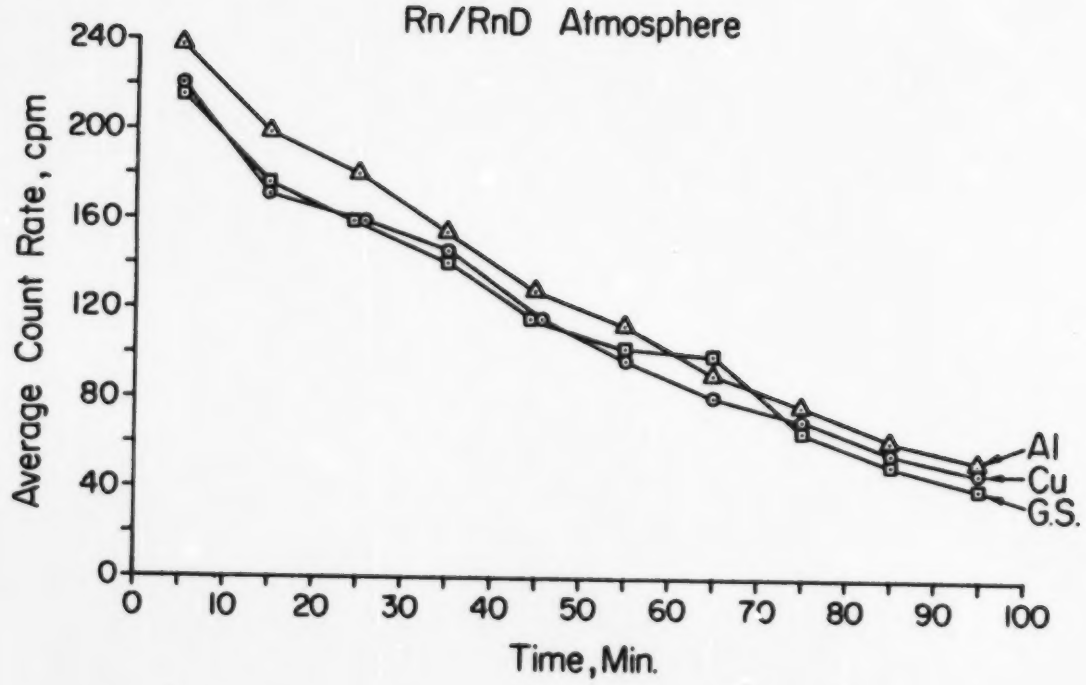


Fig. 1 - Alpha-activity versus time from radon progeny plated-out on several materials.

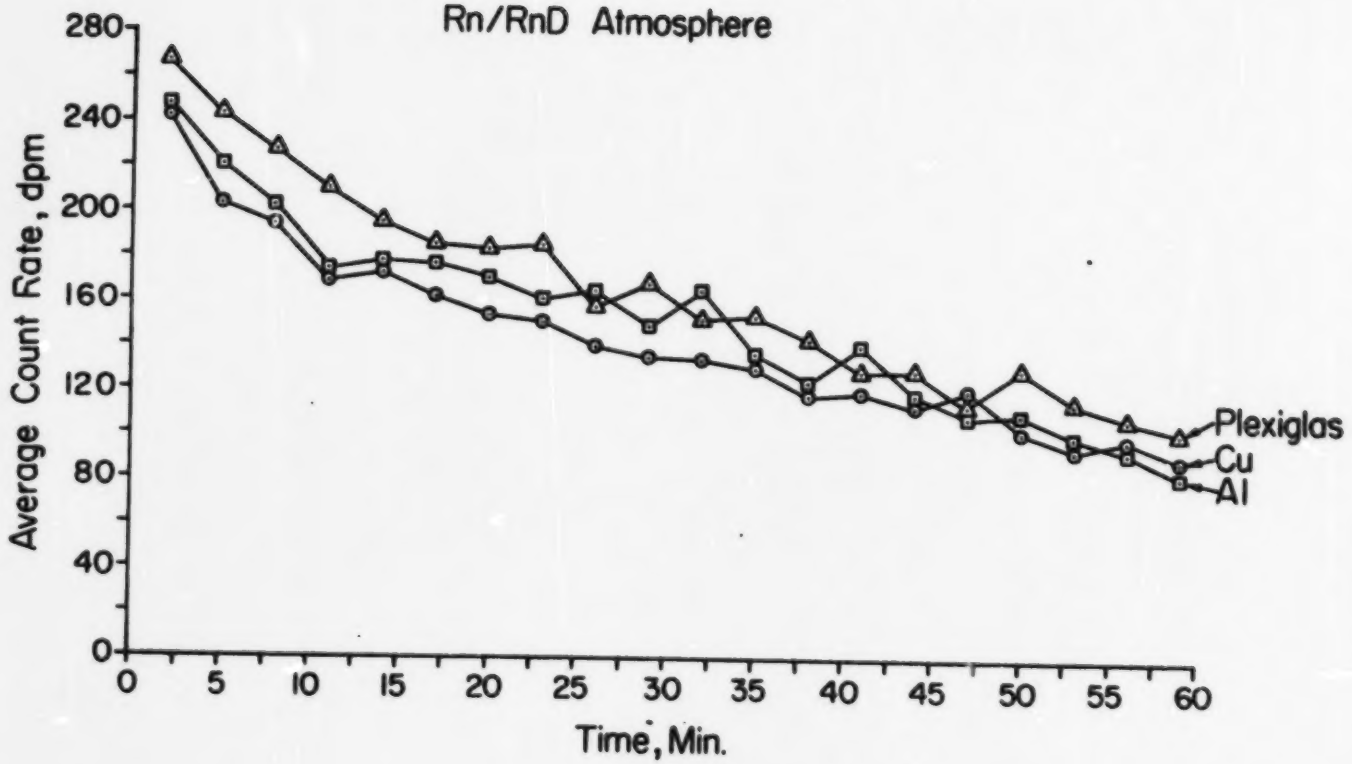


Fig. 2 - Alpha-activity versus time from radon progeny plated-out on several materials.

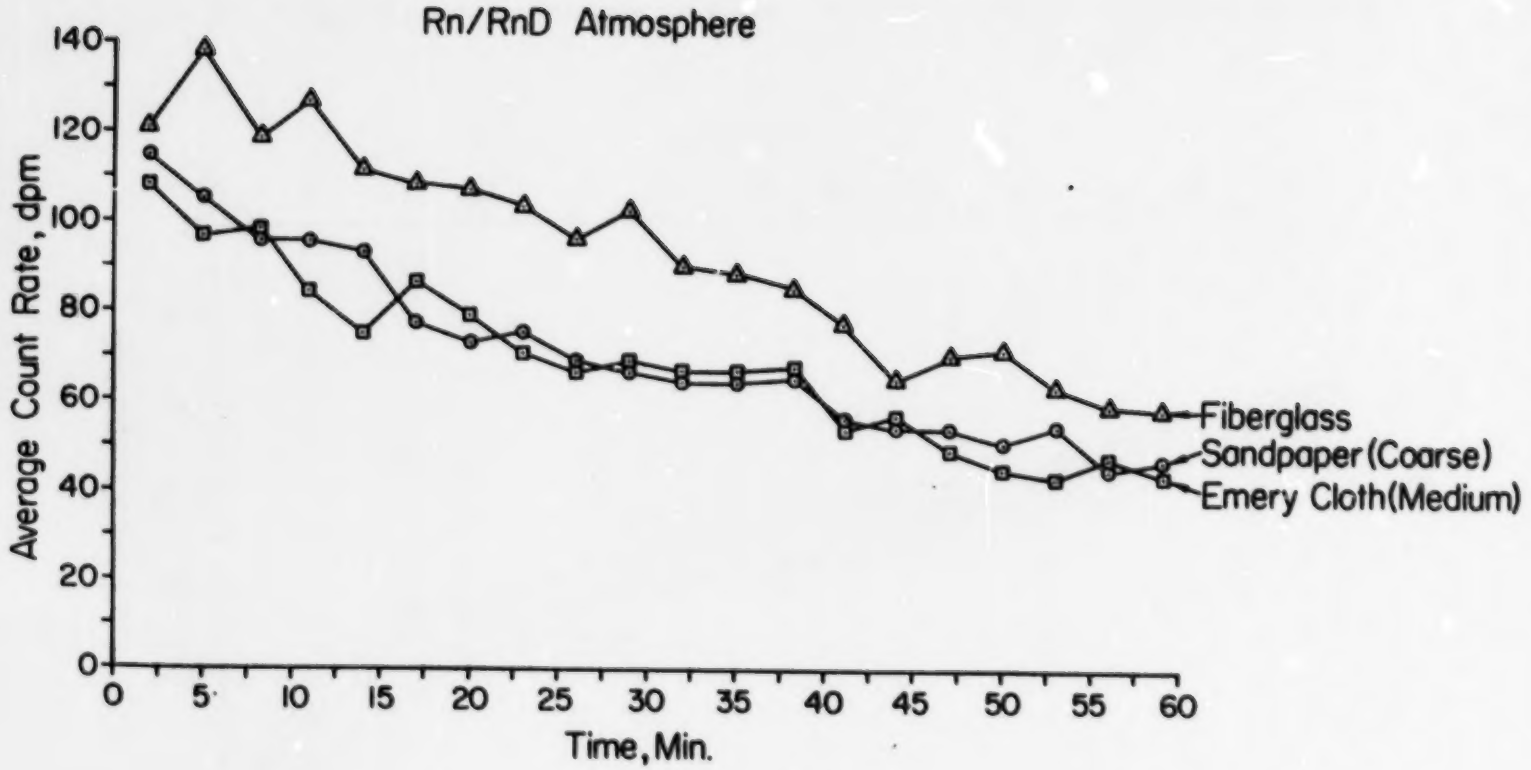


Fig. 3 - Alpha-activity versus time from radon progeny plated-out on several materials.

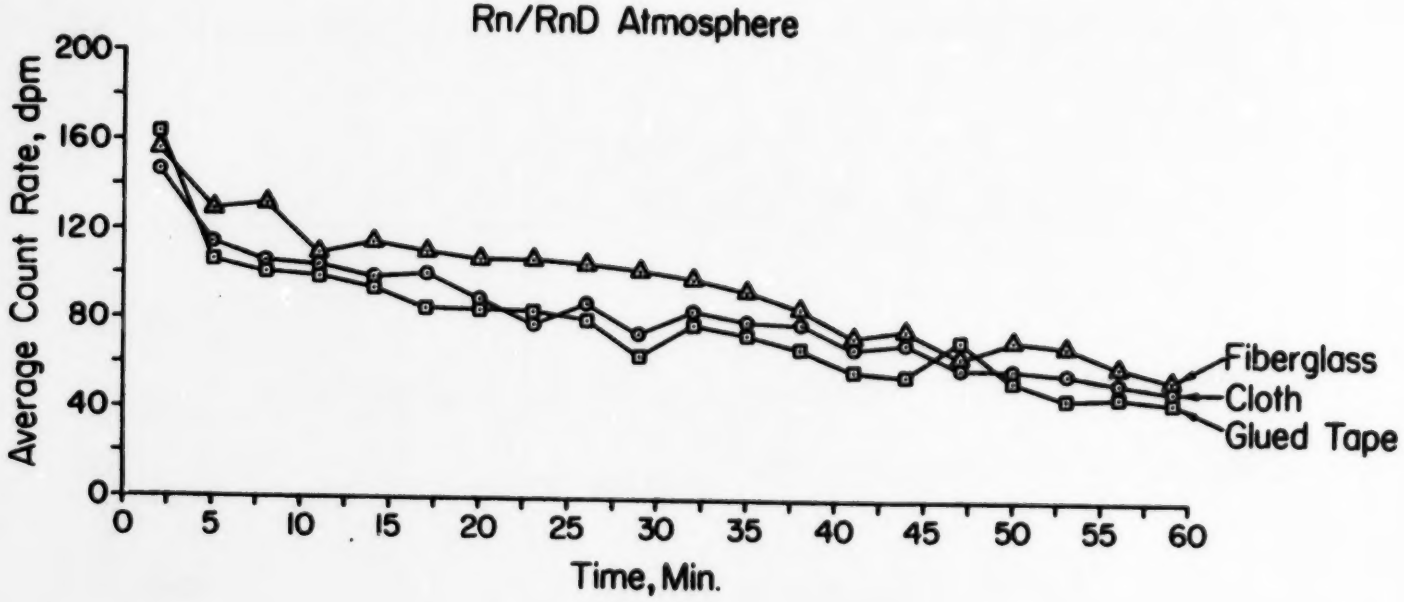


Fig. 4 - Alpha-activity versus time from radon progeny plated-out on several materials.

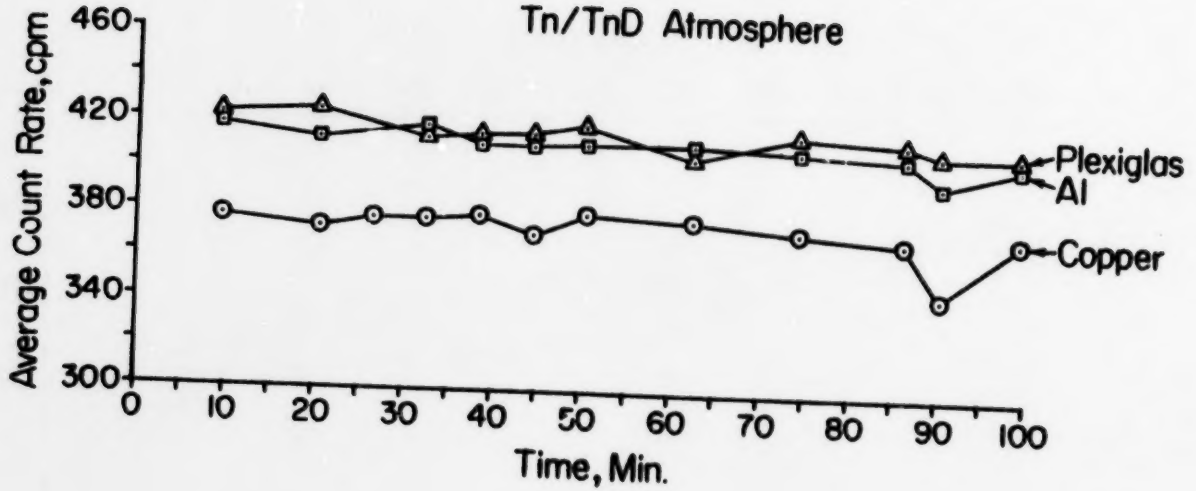


Fig. 5 - Alpha-activity versus time from thoron progeny plated-out on several materials.

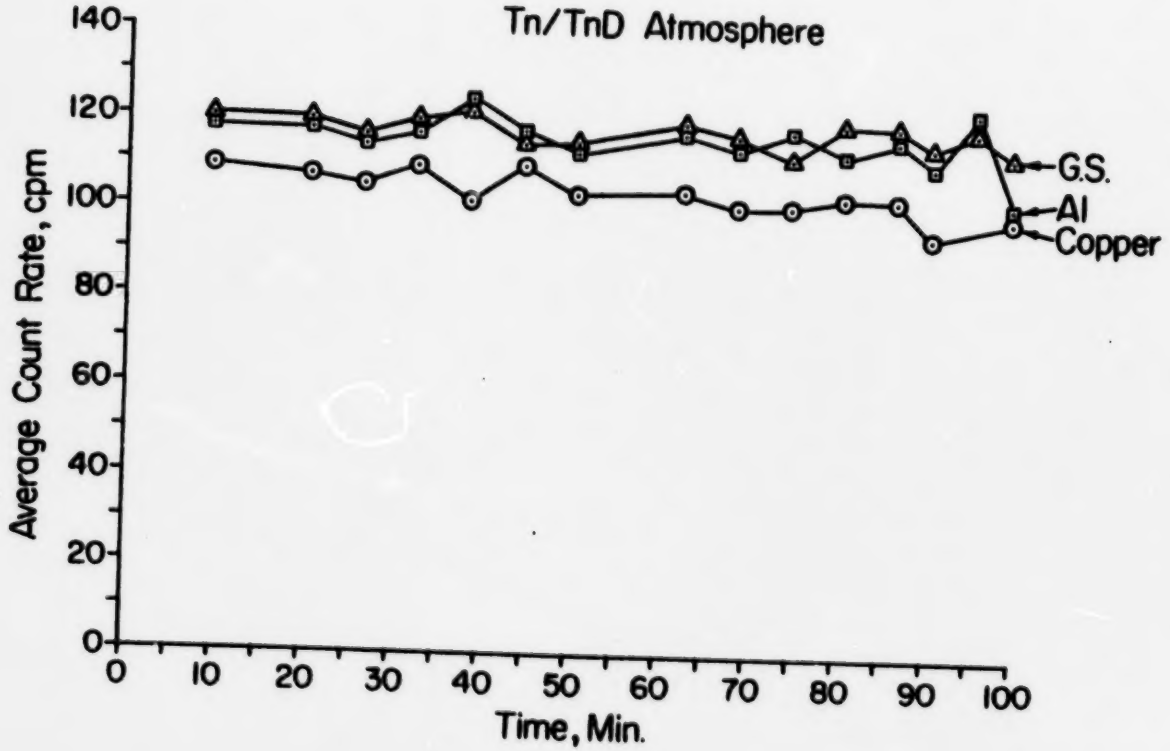


Fig. 6 - Alpha-activity versus time from thoron progeny plated-out on several materials.

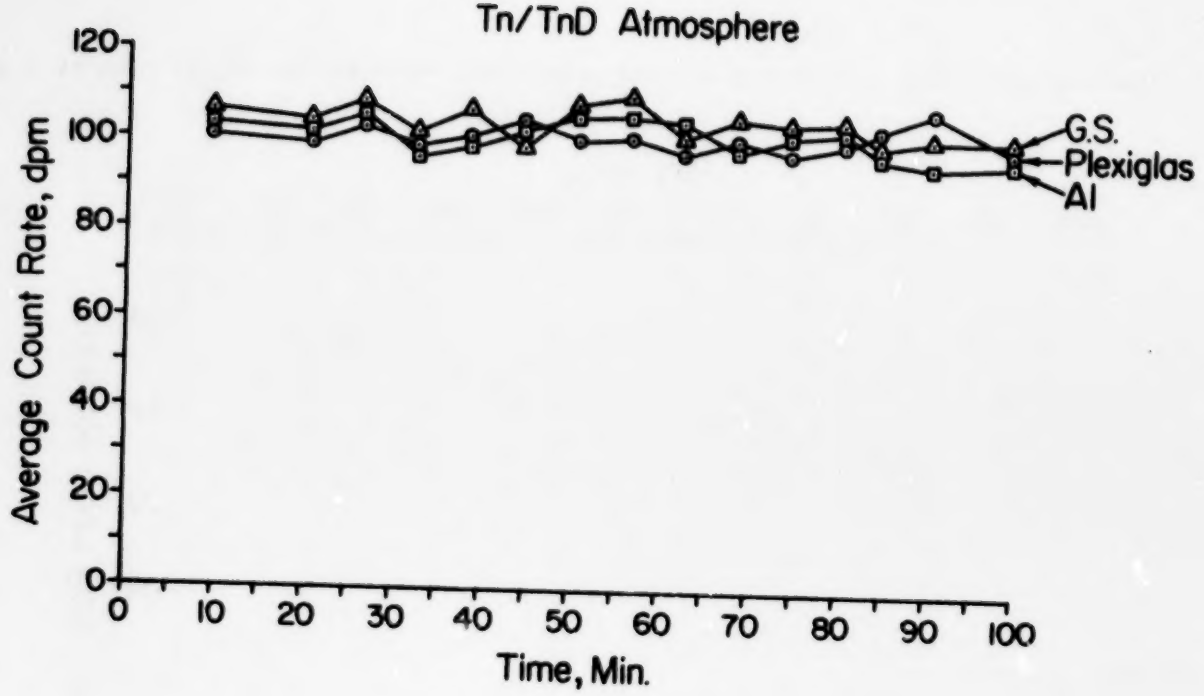


Fig. 7 - Alpha-activity versus time from thoron progeny plated-out on several materials.

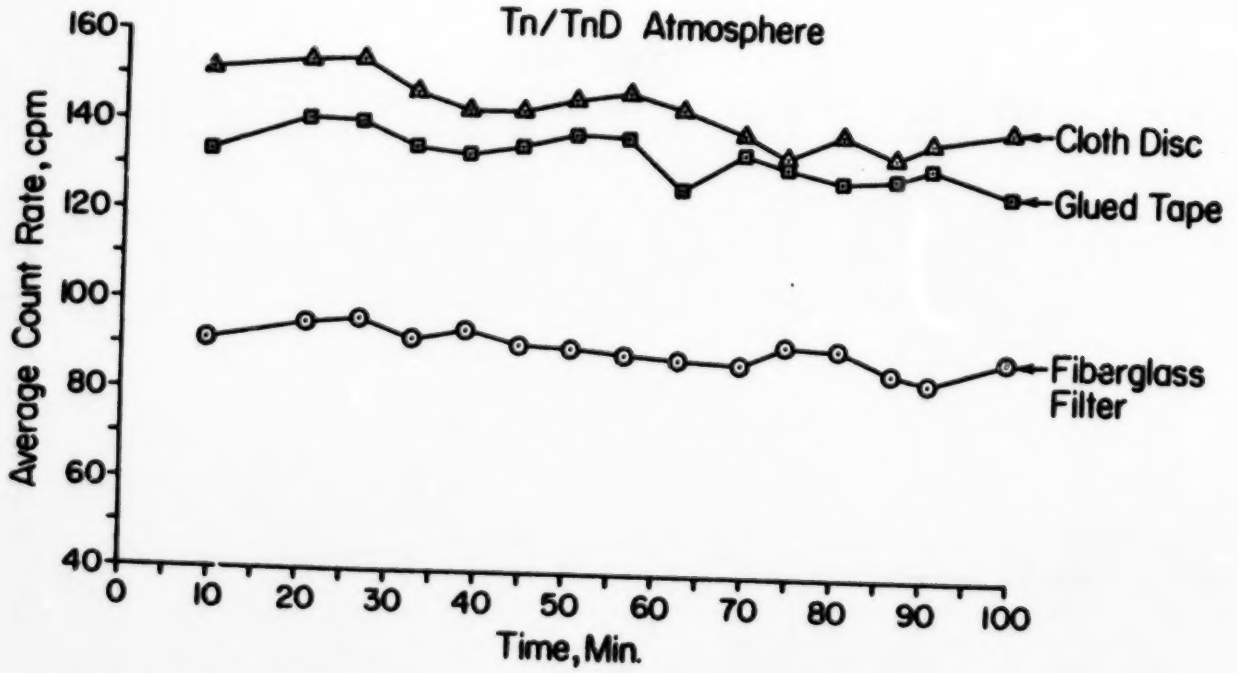


Fig. 8 - Alpha-activity versus time from thoron progeny plated-out on several materials.

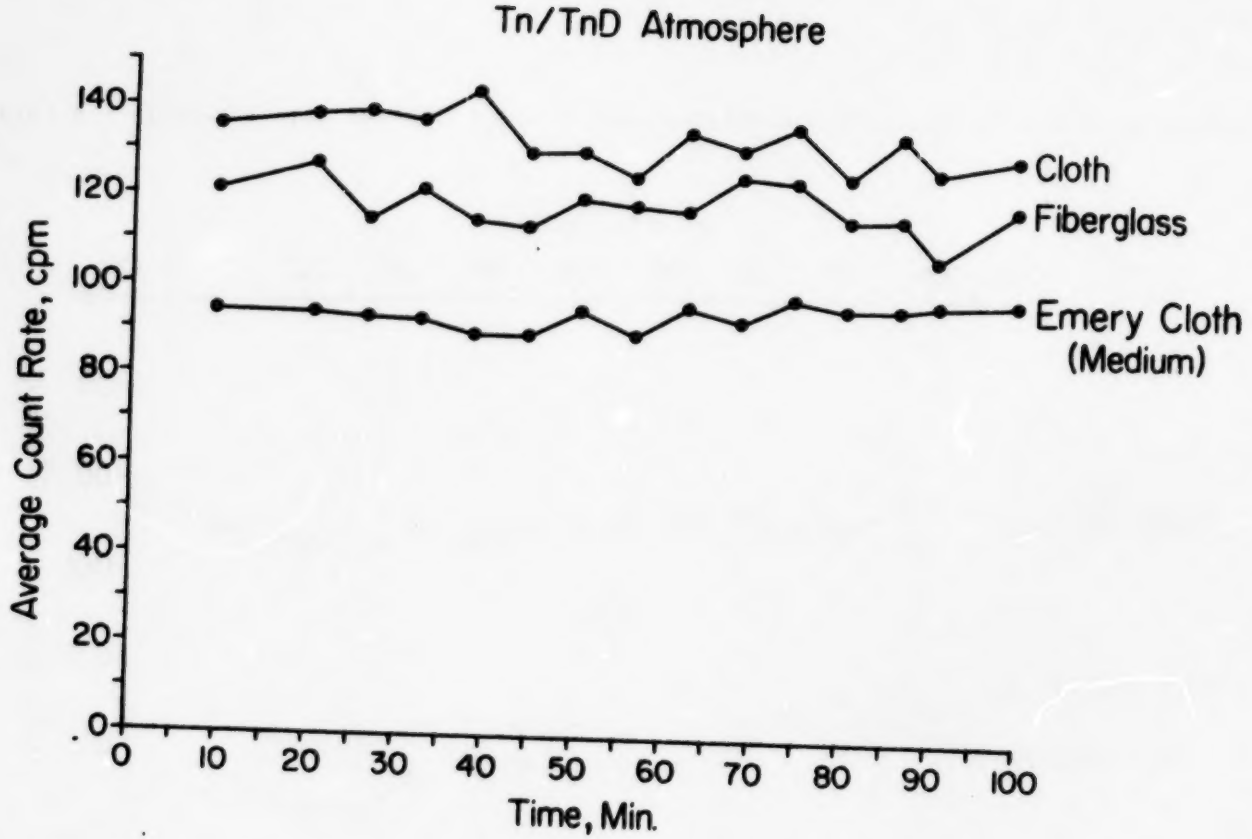


Fig. 9 - Alpha-activity versus time from thoron progeny plated-out on several materials.

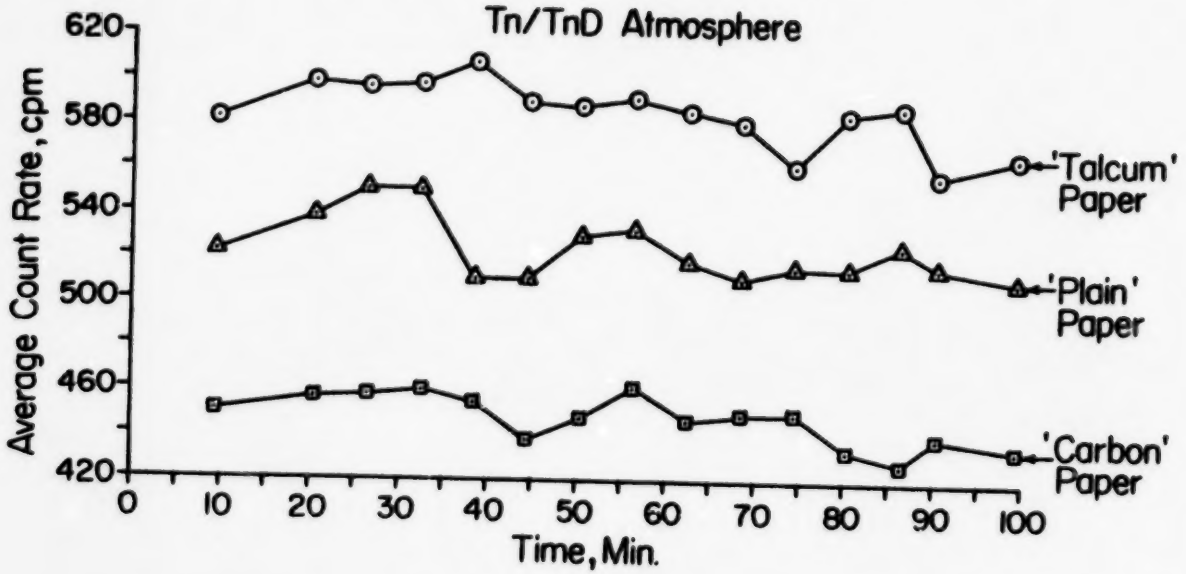


Fig. 10 - Alpha-activity versus time from thoron progeny plated-out on several materials.

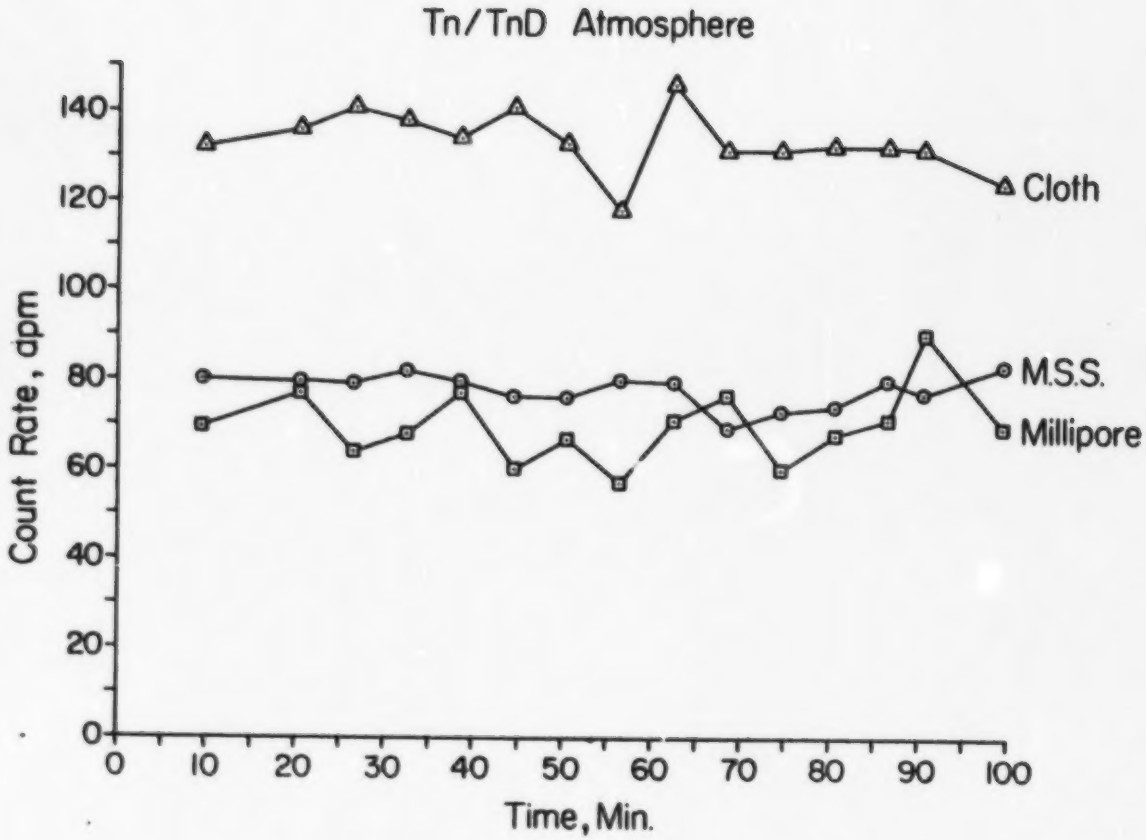


Fig. 11 - Alpha-activity versus time from thoron progeny plated-out on several materials.

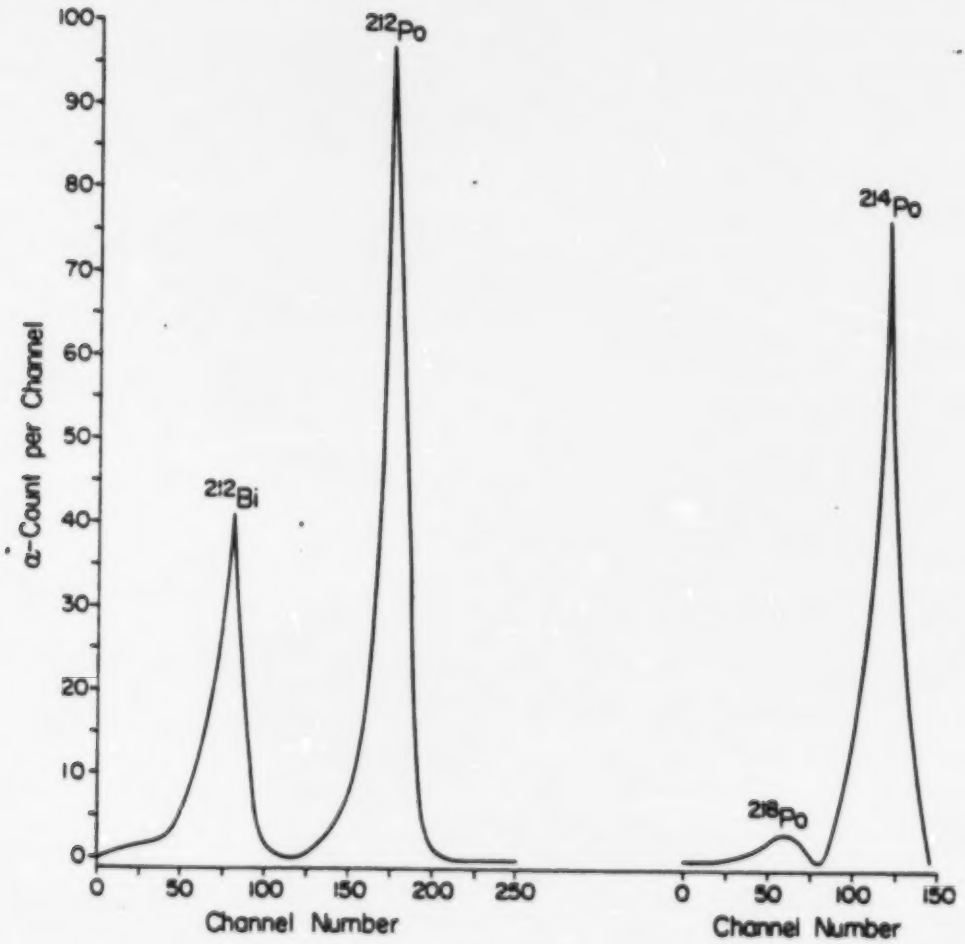


Fig. 12 - Alpha-spectra from radon progeny (right), and thoron progeny (left) plated-out on copper discs.